

Mode-Locking and Mode-Competition in a Non-equilibrium Solid-State Condensate

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A trapped polariton condensate with continuous pumping and decay is analyzed using a generalized Gross-Pitaevskii model. Whereas an equilibrium condensate is characterized by a macroscopic occupation of a ground state, here the steady-states take more general forms. Some are characterized by a large population in an excited state, and others by large populations in several states. In the latter case, the highly-populated states synchronize to a common frequency above a critical density. Estimates for the critical density of this synchronization transition are consistent with experiments.

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Recent experiments [1, 2, 3] have provided substantial evidence for a new type of Bose-Einstein condensate, formed from polaritons in semiconductor microcavities [4]. Although in many respects these results parallel those of condensation in atomic gases, the similarities conceal some fundamental differences. In particular, the lifetime of a polariton is typically only a few picoseconds, and is less than the lifetime of the condensate [2]. The condensate is therefore a non-equilibrium steady-state, in which the decaying polaritons are continually being replenished.

Several consequences of this non-equilibrium aspect of the system have now been predicted, based on both microscopic calculations and generalizations of the Gross-Pitaevskii approach [5, 6, 7, 8]. While these predictions are undoubtedly interesting, more dramatic departures from the physics of equilibrium condensates are seen experimentally. An equilibrium condensate is characterized by a macroscopic occupation at the chemical potential; for a trapped, ideal Bose gas this is the ground-state energy of the trap, while more generally it is the lowest eigenvalue of the Gross-Pitaevskii equation. In some experiments there is indeed evidence for equilibrated polariton distributions and large occupations of the $\vec{k} = 0$ ground-state of untrapped polaritons [1, 2, 3], but in others there are massive occupations associated with excited states. This has been observed in pillar traps [9], lattices [10], and disorder [11, 12]. Such results have been argued to be evidence of polariton lasing [9, 13], distinct from condensation, but this distinction is clear only to the extent that condensation is restricted to equilibrium.

The aim of this paper is to outline a theory of the trapped polariton condensate with pumping and decay. The treatment is based on a recently-proposed generalized Gross-Pitaevskii equation (gGPE) [5, 6]. Motivated by the recent experiments, we go beyond the assumption [5, 6, 7, 8, 14, 15] of condensation at a single ground-state energy, and systematically investigate how the dynamics of the pumped system can lead to more general forms. We first consider a small trap at low densities, and show that the gGPE reduces to kinetic equations for the occupations of the trap states. The occupations of the states

are determined by gain and loss processes which are independent of energy, *i.e.* a laser-like mode competition. Thus the steady-state solutions can include massive occupations of an excited state, or of several states simultaneously; the character of the steady-states in this limit can be predicted from the wavefunctions of the trap.

Having established the steady-state structure at low densities, we then numerically explore how it changes away from this limit. We demonstrate that, if the trap is such that there are several massively-occupied states at low densities, then above a threshold density the steady-state reverts to a massive occupation at a single energy. This can be understood as the classical phenomenon of synchronization (mode-locking) of coupled nonlinear oscillators [16]. Supposing that the dominant nonlinearity is the polariton-polariton interaction we predict that, for realistic parameters, polariton systems could be tuned through the synchronization transition. They thus give access to both an interaction-dominated regime, and a regime of laser-like mode competition.

Synchronization in the polariton condensate has been independently considered by Wouters [17]. That treatment is complementary to this one, considering the specific problem of a double-well trap using a related model. The double-well has also been considered under resonant pumping [18], which further differs from the present work because the condensate is directly induced by the pump.

We consider the order-parameter equation [5]

$$i\partial_t\psi = [H_0 + U|\psi|^2 + i(\gamma_{\text{eff}}(r) - \Gamma|\psi|^2)]\psi. \quad (1)$$

This complex Ginzburg-Landau form [19] has been argued [5, 6] to be the generic mean-field description of a polariton condensate with continuous pumping and decay. The first two terms on the right form the standard Gross-Pitaevskii mean-field equation, with local density $|\psi|^2$. H_0 includes a quadratic approximation to the polariton dispersion, and a potential due to disorder or the trap. $U > 0$ is the strength of the repulsive interaction between polaritons, which is treated as a contact interaction because its range is of the order of the exciton Bohr radius. The polarization degeneracy of the polaritons has been neglected for simplicity; it could be treated using a

two-component order-parameter equation [14].

The last two terms on the right of (1) account for the pumping and decay. The pumping model involves a reservoir of high-energy particles, created by some external excitation. The condensate is populated by stimulated scattering from this reservoir, contributing a linear gain term $i\gamma(r)\psi$, where $\gamma(r)$ is related to the reservoir density. This term combines with a similar term from the decay of the polaritons, giving the overall linear gain term, with coefficient γ_{eff} , in (1). However, the rate of condensate growth should reduce with increasing density, as the pump reservoir becomes depleted. This effect, modeled using a single gain-saturation coefficient Γ , gives the final term in (1). Physically, this form of pumping can be interpreted in terms of a pump which tries to locally enforce a steady-state density $\gamma_{\text{eff}}(r)/\Gamma$.

Strong-Trapping Limit– The steady-states of (1) can be determined analytically in the limit of strong trapping, where the nonlinearities are weak compared with the single-particle level spacing. We may then treat them with degenerate perturbation theory. We expand $\psi(r, t)$ in terms of the eigenstates of H_0 , ψ_n^0 , and retain only resonant terms in the resulting equations. To simplify the notation we analyze a trap with only two single-particle states, so $\psi(r, t) = \mu_1(t)\psi_1^0(r) + \mu_2(t)\psi_2^0(r)$, and assume homogeneous pumping. The amplitude $\mu_1(t)$ ($\mu_2(t)$) obeys

$$i\dot{\mu}_{1(2)} = [E_{1(2)} + i\gamma_{\text{eff}} + (U - i\Gamma) \times (\eta_{1(2)}|\mu_{1(2)}|^2 + 2b|\mu_{2(1)}|^2)] \mu_{1(2)}. \quad (2)$$

E_i is the single-particle energy, and the wavefunctions have been taken to be normalized and real. $\eta_1 = u_{1111}$, $\eta_2 = u_{2222}$, and $b = u_{1122}$ are matrix elements for a local nonlinearity,

$$u_{ppqq} = \int (\psi_p^0)^2 (\psi_q^0)^2 dr. \quad (3)$$

They parametrize the inhomogeneous density profile of the trap states, with η_1 and η_2 describing the inhomogeneity of the states, and b their overlap.

Introducing number and phase variables in a rotating frame,

$$\mu_{1(2)}(t) = e^{-i\gamma_{\text{eff}}U/\Gamma} \sqrt{n_{1(2)}(t)} e^{-i\phi_{1(2)}(t)}, \quad (4)$$

separates the number and phase dynamics. The former obeys the rate equations

$$\dot{n}_{1(2)} = 2\Gamma n_{1(2)} (\gamma_{\text{eff}}/\Gamma - \eta_{1(2)} n_{1(2)} - 2bn_{2(1)}), \quad (5)$$

with terms describing the stimulated scattering from the reservoir and the spontaneous decay, and the reservoir depletion. This result can be understood as a generalization of the kinetic description of polariton lasing [20, 21, 22, 23], to treat the spatial structure of the trap.

It describes the extensive component of the occupation, and hence effects such as spontaneous pumping are missing. They would be important for a finite system close to threshold [24].

The phase dynamics is straightforward, obeying

$$\dot{\phi}_{1(2)} = E_{1(2)} - U(\gamma_{\text{eff}}/\Gamma - \eta_{1(2)} n_{1(2)} - 2bn_{2(1)}). \quad (6)$$

Each mode oscillates freely, at a single-particle energy which is shifted by the repulsive interactions.

The rate equations (5) have several steady-state solutions above the bulk condensation threshold, $\gamma_{\text{eff}} = 0$. There are always two solutions corresponding to condensation in each of the trap states: either $n_1^s = \gamma_{\text{eff}}/(\eta_1\Gamma)$ is finite and $n_2^s = 0$ vanishes (state S_1), or *vice versa* (state S_2). However, if either $\eta_1, \eta_2 > 2b$ or $\eta_1, \eta_2 < 2b$ there is also a steady-state T , with massive occupations of both trap states:

$$n_{1(2)} = \frac{\gamma_{\text{eff}}}{\Gamma} \left(\frac{2b - \eta_{2(1)}}{4b^2 - \eta_1\eta_2} \right). \quad (7)$$

The left panel of Fig. 1 shows the parameter regions in which the different steady-states are stable. Linearizing (5) about the steady-state S_1 , we find that population fluctuations decay with rates $\lambda_1 = 2\gamma$ and $\lambda_2 = 2\gamma(2b/\eta_1 - 1)$. Thus this solution is stable for $\eta_1 < 2b$. This is the condition that the occupation of the first trap state alone, determined by the pumping and the self-gain-saturation parameter η_1 , is sufficient to keep the second below threshold. The analogous condition $\eta_2 < 2b$ holds for the existence of a stable solution in which only the second trap state is occupied, S_2 . If neither criterion is satisfied both states must be occupied, and this is where the two-mode solution T is stable – fluctuations there decay with rates 2γ and $2\gamma(2b - \eta_1)(2b - \eta_2)/(4b^2 - \eta_1\eta_2)$.

The different steady-state solutions can be realized in many different potentials. For two widely-separated states we generically have $b \rightarrow 0$, and hence obtain two coexisting condensates. While this result is perhaps expected for separated traps, it can nonetheless also occur when there is substantial spatial overlap. A simple model which demonstrates this is a one-dimensional hard-wall trap of size a , with a finite trap of size b and depth V at its center. The right panel of Fig. 1 shows the different steady-state regions for the two lowest eigenstates of this potential.

Experimentally, the presence of massively-occupied states in polariton systems is shown by bright luminescence peaks. The characteristic frequencies in such optical spectra follow from (6). In $S_1(S_2)$ the massively-occupied states will lead to strong emission at the frequency $U\gamma_{\text{eff}}/\Gamma + E_{1(2)}$, while in T both peaks appear simultaneously. Note that here the blueshift of the condensing modes is $U\gamma_{\text{eff}}/\Gamma$, irrespective of which steady-state we consider. Physically, this is because the energy shifts are determined by the density, which is fixed by the pumping.

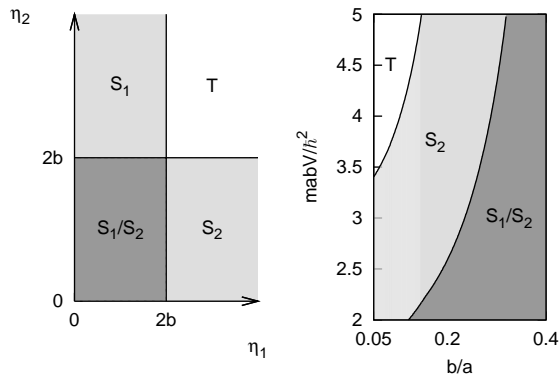


FIG. 1: Left panel: steady-state structure for condensation in a two-state trap, for the generalized Gross-Pitaevskii model neglecting non-resonant terms (2). Lettering denotes the stable steady-states in each region, with condensation in both trap states (T), state 1 alone (S_1), or state 2 alone (S_2). In the lower-left region both S_1 and S_2 are stable, and the steady-state is selected by the initial conditions. Right panel: steady-state structure for the two lowest states of a one-dimensional model trap, consisting of a hard-wall trap of length a with a trap of length b and depth V at its center.

In addition to the strong emission associated with the condensing states, we also expect peaks in the optical response associated with the non-condensing trap states. For the mean-field model in the resonant approximation these states are not populated, and hence would appear in the absorption but not the luminescence. However, they could develop non-macroscopic populations due to effects beyond that model, in which case they would appear weakly in luminescence. These peaks are shifted by the mean-field repulsion with the condensate, so in S_1 the absorption peak will be at $E_2 + 2bn_1$. Since its width will be proportional to λ_2 , the mode is narrow close to the critical line $\eta_1 = 2b$, where it goes unstable.

Beyond Strong-Trapping— Having established the physics of (1) in the strong-trapping regime, we now investigate how it develops with increasing nonlinearity. To do this we use real-space discretization to solve (1) directly, for the one-dimensional model described above. This potential is chosen as a simple, few-parameter model, which can be tuned into the different steady-state regions. We choose $a = m = \Gamma = 1$, taking a as the unit of length, $\hbar^2/(ma^2)$ as the unit of energy, and working with a rescaled density $|\psi|^2 \rightarrow |\psi|^2/\Gamma$. Both the repulsive interaction U and the gain saturation Γ are believed to be significant for polaritons [1, 5, 6], and so for this demonstration we take $U/\Gamma = 1$.

Fig. 2 illustrates how the steady-state spectra develop with increasing pumping. The analysis above, applied to the two lowest states of this potential, predicts the two-mode steady-state T . This is in agreement with the results at weak pumping (lowest panel). Increasing the pumping there is the overall blueshift associated

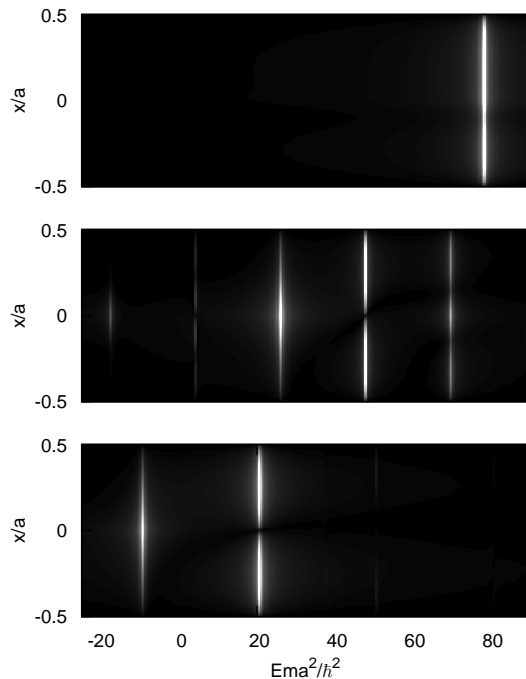


FIG. 2: Spectral analysis of the polariton field in the steady-states of the generalized Gross-Pitaevskii model, with the potential described in the caption of Fig. 1. The pump strength increases through $\gamma_{\text{eff}} = 1, 30, 60$ from the lowest panel to the highest. The grayscale is the computed amplitude $|\psi(E, x)|$, normalized by $\sqrt{\gamma_{\text{eff}}}$ to account for the overall increase in density. $U/\Gamma = 1$, $b = 0.05$ and $V = 25$.

with the increased density. The lowest two modes still dominate the spectrum, but their energy splitting has reduced slightly, and several further emission peaks appear (middle panel). Further increasing the pumping, the spectrum switches to emission at a single frequency (top panel).

At a general level, these results are expected consequences of the non-resonant terms dropped from (2). In a two-mode model with states of opposite parity, for example, there is the additional Josephson term

$$b(U - i\Gamma)\mu_{1(2)}^*\mu_{2(1)}\mu_{2(1)} \quad (8)$$

in the equation (2) for $\mu_1(\mu_2)$. Thus the condensing states can drive nonlinear emission at other frequencies, as we see at intermediate pumping. Furthermore, the non-resonant terms couple together the phases of the condensing modes in (6). Thus we expect frequency pulling, and eventually synchronization of the condensing states [16].

Coupled oscillators synchronize to a common frequency when the phase-phase couplings become comparable to their energy splitting Δ . Since here the states have similar and overlapping density profiles, the matrix elements (3) are all of order one. The scale of the phase-dependent couplings in (6) is then set only by the non-

linearity $\sqrt{U^2 + \Gamma^2}$ and the polariton density $\rho \approx \gamma_{\text{eff}}/\Gamma$. We may thus estimate the critical polariton density for the mode-locking transition, ρ_c , from

$$\Delta \sim \rho_c \sqrt{U^2 + \Gamma^2} \sim \gamma_{\text{crit}} \sqrt{1 + U^2/\Gamma^2}. \quad (9)$$

This form is consistent with Fig. 2, and with simulation results (not shown) for other values of the nonlinearities U and Γ . In particular, in the same model the synchronization occurs between $\gamma_{\text{eff}}/\Gamma = 20$ and 40 when $U = 0$, and between $U/\Gamma = 20$ and 40 at fixed $\gamma_{\text{eff}}/\Gamma = 1$.

Synchronization due to nonlinear gain is well-known as the basis for mode-locked lasers. However, an important difference between the polariton condensate and a laser is the presence of strong interactions U between the particles, due to the excitonic component of the polariton. Thus frequency-pulling and synchronization could be expected to have a much wider role in the physics of polariton condensates than they do in lasers, occurring on large energy scales at low intensities.

Let us estimate the critical polariton density ρ_c for synchronization, supposing it is controlled by the real nonlinearity U . Estimates of this interaction are available for plane-wave excitons in a perfect quantum-well, and the localized exciton states of a disordered quantum well [25]. For current experiments, a plausible upper limit is the result $U \sim \Omega_R(m_x W)^{-1}$ from the disordered models, with disorder energy-scale $W \sim 1\text{meV}$. $m_x \sim 0.5m_e$ is the exciton mass, and $\Omega_R \sim 20\text{meV}$ the Rabi splitting. Thus the phase boundary for synchronization in a trap of scale L_t is

$$\Omega_R(m_x W)^{-1} \rho_c \sim 1/(m L_t^2). \quad (10)$$

For a trap of $L_t \sim 5\mu\text{m}$ [9] and a polariton mass $m \sim 10^{-5}m_e$ this gives $\rho_c \sim 10^{11}\text{cm}^{-2}$, or $n_c \sim L_t^2 \rho_c \sim 10^3$. This estimate is about one order of magnitude larger than the densities usually reported for polariton condensates. Thus, with a suitable potential, coexisting polariton condensates of different frequencies should be expected in tight traps. The synchronization transition could then be observed with increasing density, if necessary using larger traps to reduce ρ_c into the experimentally accessible range.

Since we do not expect synchronization at current densities in tight traps, the present theory is consistent with the observations there of well-resolved emission lines [9, 12]. It also appears to be consistent with the existence of long-range coherence [1, 11] at current densities, with softer traps formed by disorder.

Concluding Remarks—Because the tightly-trapped polariton condensate has well-resolved emission lines, it could provide a sensitive probe of the physics of non-equilibrium condensates. In particular, the present results will allow the order-parameter equation (1) to be tested. While this form certainly captures much of the physics [5, 6], effects which are important elsewhere are

missing. Most obviously, there is no thermalization with the reservoirs [26, 27], which exchange particles with the system irrespective of energy, and do not directly cause transitions between trap states. Such effects would appear in generalizations of the kinetic equations (5). Since some groups [1, 2] report thermalized distributions, such generalizations may prove necessary.

To conclude, we have considered the trapped polariton condensate in the framework of a generalized Gross-Pitaevskii model. At low densities, this model admits solutions which, differently from an equilibrium condensate, involve massive occupations of excited states, or of several states simultaneously. We have derived criteria for predicting the nature of the steady-states in a given geometry, and shown that the steady-states are selected by gain-competition effects. At a general level such physics is of course familiar in lasers, though it has not previously been considered for the polariton system. Moreover, the direct interactions between polaritons create differences compared with the photon laser: blueshifting the modes in the weak-nonlinearity regime, and causing frequency-pulling and synchronization at stronger nonlinearities. Our estimates for the critical density of the synchronization transition suggest that it could be cleanly observed in tight traps [9], and may be responsible for the observations of long-range coherence across disorder potentials [1, 11].

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